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Applicants: Hiroshi YOKOYAMA et al.

Serial No.: 10/539,054

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For: POLYTRIMETHYLENE TEREPHTHALATE RESIN AND  
METHOD FOR PRODUCING THE SAME

Art Unit: 4134

Examiner: Thomas Matochik

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DECLARATION UNDER 37 C.F.R. 1.132

I, the undersigned, Hiroshi YOKOYAMA, a Japanese citizen, residing at Michel-Fuji 202, 117-1, Mitojima, Fuji-shi, Shizuoka-ken 700-0913 Japan, hereby declare and state that:

I took a master course at the Division of Applied Precision Chemistry, School of Engineering, Osaka University, and I was graduated therefrom in March 1989.

I entered Asahi Kasei Kogyo Kabushiki Kaisha in April 1989.

I have been engaged in the research and development of high performance polyester resins from April 1989 to date.

I am one of the applicants of the above-identified application and I am well familiar with the present case.

I have read and understood the Office Action dated October 31, 2007 and the references cited therein.

I carried out Production Examples 1 to 14, Examples 1 to 24, Comparative Examples 1 to 17 of the present application, and the results are as described on pages 157 to 217 of the specification of the present application.

I have made observations, with reference to Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification, to show that any of the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application, is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application, with reference to Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification. The method and results are as described in a paper attached hereto and marked "Exhibit 1".

From the results of Exhibit 1, it can be fairly concluded:

that, as shown in Table A in Exhibit 1, each of Examples 1 and 2 satisfies all requirements of the method of claim 3 of the present application;

that on the other hand, as also shown in Table A in Exhibit 1, each of Comparative Examples 1 and 4 employs a crude PTT resin having an E value of 0.071; that is, Comparative Examples 1 and 4 do not satisfy the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application;

that further, as also shown in Table A in Exhibit 1, Comparative Example 9 employs a *solid-phase* polymerization process; that is, Comparative Example 9 does not satisfy the "in a molten form" requirement of step (2) of the method of claim 3 of the present application;

that, as indicated in Table B in Exhibit 1, the results of Examples 1 and 2 (i.e. the properties of the PTT resins obtained) are excellent; specifically, the PTT resins obtained satisfy all requirements of claim 1 of the present application;

that, as also indicated in Table B in Exhibit 1, the results of Comparative Examples 1 and 4 (i.e. the properties of the PTT resins obtained) are poor; specifically, the PTT resins obtained in Comparative Examples 1 and 4, respectively,

exhibit cyclic dimer contents (% by weight) of 2.45 and 2.34, which do not satisfy the cyclic dimer content (% by weight) requirement (not greater than 2 % by weight) of claim 1 of the present application;

that, as also indicated in Table B in Exhibit 1, the results of Comparative Example 9 (i.e. the properties of the PTT resin obtained) are poor; specifically, the PTT resin obtained exhibits a molecular weight distribution (Mw/Mn) of 3, which does not satisfy the molecular weight distribution (Mw/Mn) requirement (from 2 to 2.7) of claim 1 of the present application; and also the PTT resin obtained exhibits a crystallinity as high as 55 (which causes high brittleness);

that, thus, a comparison between Examples 1 and 2 and Comparative Examples 1 and 4 clearly shows that the polytrimethylene terephthalate resin of the present invention cannot be obtained when the crude PTT resin used does not satisfy the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application; in other words, the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application;

that, further, a comparison between Examples 1 and 2 and

Comparative Example 9 clearly shows that the polytrimethylene terephthalate resin of the present invention cannot be obtained by a solid-phase polymerization process that is a representative conventional technology; in other words, the "in a molten form" requirement of step (2) of the method of claim 3 of the present application is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application;

that Comparative Example 9 also shows that, even in the case where there are satisfied the requirements of step (1) of the method of claim 3 (including the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3), the excellent PTT resin of the present invention cannot be obtained when the "in a molten form" requirement of step (2) of the method of claim 3 is not satisfied;

that thus, Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification clearly show that the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application can be obtained only when there are satisfied both the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the

present application; and

that therefore, it is quite apparent that any of the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application, is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application.

The undersigned petitioner declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: February 12, 2008

Hiroshi Yokoyama  
Hiroshi YOKOYAMA

Exhibit 1

Observations to show that any of the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application, is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application, with reference to Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification

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1. The object of observations

The polytrimethylene terephthalate (PPT) resin of the present invention not only has a low cyclic dimer content but is also capable of suppressing the formation of the cyclic dimer even during the melt molding, thereby enabling the production of an excellent shaped article stably on a commercial scale, the shaped article having advantages not only in that the shaped article has high strength and excellent color, but also in that the shaped article is free from the bleeding of the cyclic dimer to the surface of the shaped article, so as to be suitable for coating with a coating composition or adhesive agent and exhibit excellent adhesion property. By the method of claim 3 of the present application, the excellent polytrimethylene terephthalate (PPT) resin of the present in-

vention can be produced stably with high productivity on a commercial scale.

The object of the present observations is to show that any of the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application, is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application.

## 2. Observations

The present invention exhibits the above-described excellent effects, as compared to the conventional melt polymerization processes and the solid-phase polymerization processes as well as the conventional polytrimethylene terephthalate (PPT) resin produced by the prior art processes.

Such excellent effects of the present invention are fully substantiated by Examples 1 to 24 and Comparative Examples 1 to 17 of the present specification. (Most important data of Examples 1 to 24 and Comparative Examples 1 to 17 are indicated in Tables 1 to 5 at pages 211 to 217 of the present specification.)

Especially, attention is drawn to Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification.

As described hereinbelow, a comparison between Examples



1 and 2 and Comparative Examples 1 and 4 clearly shows that the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application is critical for producing the excellent polytrimethylene terephthalate resin of the present invention.

As also described hereinbelow, a comparison between Examples 1 and 2 and Comparative Example 9 clearly shows that the polytrimethylene terephthalate resin of the present invention cannot be obtained by a solid-phase polymerization process that is a representative conventional technology (employed alone or in combination with a conventional melt polymerization process). In other words, the "in a molten form" requirement of step (2) of the method of claim 3 of the present application is critical for producing the excellent polytrimethylene terephthalate resin of the present invention.

The most important data of Examples 1 and 2 and Comparative Examples 1, 4 and 9 are indicated in Table 2 at pages 212 and 213 of the present specification. For easier reference, the data of Examples 1 and 2 and Comparative Examples 1, 4 and 9 (as in Table 2 of the present specification) are collected and shown in Tables A and B below. (The data collected from "Table 2 (to be continued)" at page 212 is indicated in Table A, and the data collected from "Table 2 (continued)" at page 213 is indicated in Table B.)

Table A

	Production of crude PTT resin	Properties of crude PTT resin (prepolymer)		E value (cyclic dimer formation index)	Polymerization conditions				State of crude PTT resin in polymerizer	
		Intrinsic viscosity $[\eta]$			Mode of Polymerization (guide)	Temperature (°C)	Pressure (Pa)	Nitrogen (mg/g)	Foaming	Staining
Example 1 (present invention)	Production Example 1	0.65		0.005	Wire (melt polymerization)	255	20	0	good	small
Example 2 (present invention)	Production Example 2	0.71		0.016	Wire (melt polymerization)	255	20	0	good	small
Comparative Example 1	Production Example 4	0.72		0.071	Wire (melt polymerization)	255	20	0	good	small
Comparative Example 4	Production Example 8	1.02		0.071	Wire (melt polymerization)	250	150	0	good	small
Comparative Example 9	Production Example 1	0.65		0.005	Solid-phase polymerization	205	20	0	-	-

Table B

	Properties of final PTT resin					
	Intrinsic viscosity [ $\eta$ ]	Molecular weight distri- bution (Mw/Mn)	Cyclic dimer content (wt %)	Color		Crystallin- ity (%)
				b*	L*	
Example 1 (present invention)	1.10	2.2	1.50	1	88	5
Example 2 (present invention)	1.21	2.3	1.68	5	90	6
Comparative Example 1	1.19	2.3	2.45	7	88	5
Comparative Example 4	1.45	2.4	2.34	10	86	4
Comparative Example 9	0.78	3.0	1.01	2	88	55

As shown in Table A above, each of Examples 1 and 2 satisfies all requirements of the method of claim 3 of the present application.

On the other hand, as also shown in Table A, each of Comparative Examples 1 and 4 employs a crude PTT resin having an E value of 0.071; that is, Comparative Examples 1 and 4 do not satisfy the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application.

Further, as also shown in Table A, Comparative Example 9

employs a *solid-phase* polymerization process; that is, Comparative Example 9 does not satisfy the "in a molten form" requirement of step (2) of the method of claim 3 of the present application.

(Evaluation of the results of Examples 1 and 2 and Comparative Examples 1, 4 and 9)

As indicated in Table B above, the results of Examples 1 and 2 (i.e. the properties of the PTT resins obtained) are excellent; specifically, the PTT resins obtained satisfy all requirements of claim 1 of the present application. The results of Examples 1 and 2 are described in the present specification as follows:

(Example 1)

"The obtained PTT resin had a high molecular weight, a narrow molecular weight distribution, a low cyclic dimer content, and excellent color."  
(emphasis added) (see page 185, lines 7 to 9 of the present specification); and

(Example 2)

" In each of Examples 2 to 7, the obtained PTT resin (in the form of pellets) had a high molecular weight, a narrow molecular weight distribution, a low cyclic dimer content and excellent color."  
(emphasis added) (see page 186, lines 16 to 19 of the present specification).

As also indicated in Table B above, the results of Comparative Examples 1 and 4 (i.e. the properties of the PTT resins obtained) are poor; specifically, the PTT resins obtained in Comparative Examples 1 and 4, respectively, exhibit cyclic dimer contents (% by weight) of 2.45 and 2.34, which do not satisfy the cyclic dimer content (% by weight) requirement (not greater than 2 % by weight) of claim 1 of the present application. The results of Comparative Examples 1 and 4 are described in the present specification as follows:

" In Comparative Example 1, the crude PTT resin used as a prepolymer had a cyclic dimer formation index (E) as high as 0.071, and, hence, a PTT resin having a low cyclic dimer content could not be obtained.

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.....  
In Comparative Example 4, the crude PTT resin produced in Production Example 8 was used as a prepolymer, which crude PTT resin had a cyclic dimer content as low as 0.92 % by weight. However, the prepolymer had a cyclic dimer formation index (E) as high as 0.071, so that the cyclic dimer content of the final PTT resin could not be decreased and, on the contrary, was increased. "  
(emphasis added) (see page 187, lines 3 to 6 and 17 to 24 of the present specification).

As also indicated in Table B above, the results of Com-

parative Example 9 (i.e. the properties of the PTT resin obtained) are poor; specifically, the PTT resin obtained exhibits a molecular weight distribution (Mw/Mn) of 3, which does not satisfy the molecular weight distribution (Mw/Mn) requirement (from 2 to 2.7) of claim 1 of the present application. Also the PTT resin obtained exhibits a crystallinity as high as 55 (which causes high brittleness). The results of Comparative Example 9 are described in the present specification as follows:

" The obtained pellets were analyzed, and as a result, it was found that the obtained PTT resin had a high molecular weight, a low cyclic dimer content and good color. However, the PTT resin had a broad molecular weight distribution. Further, the pellets obtained by the solid-phase polymerization process not only had attached thereto polymer powder in an amount as large as 1 % by weight, but also had a crystallinity as high as 55 %, so that the obtained pellets were brittle. If it is attempted to transfer the obtained pellets by means of a feeder or a pneumatic conveyer, the pellets would be broken, thereby forming a large amount of polymer powder." (emphasis added) (see page 194, line 16 to 195, line 3 of the present specification).

### 3. Conclusion

Thus, a comparison between **Examples 1 and 2 and Comparative Examples 1 and 4** clearly shows that the polytrimethylene terephthalate resin of the present invention cannot be obtained when the crude PTT resin used does not satisfy the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application. In other words, the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application.

Further, a comparison between **Examples 1 and 2 and Comparative Example 9** clearly shows that the polytrimethylene terephthalate resin of the present invention cannot be obtained by a solid-phase polymerization process that is a representative conventional technology. In other words, the "in a molten form" requirement of step (2) of the method of claim 3 of the present application is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application.

Also, the following should be noted. As shown in Table

A above, the crude PTT resin (obtained in Production Example 1) used in Comparative Example 9 satisfies the requirements of step (1) of the method of claim 3. Especially, the crude PTT resin (obtained in Production Example 1) used in Comparative Example 9 has an E value of 0.005, which satisfies the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3 of the present application. Therefore, Comparative Example 9 also shows that, even in the case where there are satisfied the requirements of step (1) of the method of claim 3 (including the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) of the method of claim 3), the excellent PTT resin of the present invention cannot be obtained when the "in a molten form" requirement of step (2) of the method of claim 3 is not satisfied.

Thus, Examples 1 and 2 and Comparative Examples 1, 4 and 9 of the present specification clearly show that the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application can be obtained only when there are satisfied both the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application.



Therefore, it is quite apparent that any of the cyclic dimer formation index (E) requirement (i.e., less than 0.066) of step (1) and the "in a molten form" requirement of step (2) of the method of claim 3 of the present application, is critical for producing the polytrimethylene terephthalate resin of the present invention that satisfies all excellent features of claim 1 of the present application.